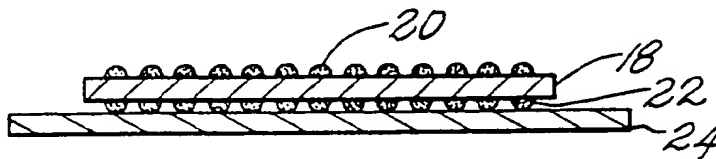


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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(21) International Application Number: PCT/US93/09996 (22) International Filing Date: 19 October 1993 (19.10.93) (30) Priority data: 07/963,674 20 October 1992 (20.10.92) US (71) Applicant: AVERY DENNISON CORPORATION [US/ US]; 150 N. Orange Grove Boulevard, Pasadena, CA 91109 (US). (72) Inventors: VARGAS, Richard, R. ; 11724 Garfield Avenue, Southgate, CA 90280 (US). PLAMTHOTTAM, Sebas- tian, S. ; 1496 Moonridge Court, Upland, CA 91786 (US). LANDERS, John, O. ; 2610 Bernwood Street, Du- arte, CA 91010 (US).		(74) Agent: GRINNELL, John, P.; Christie, Parker & Hale, P.O. Box 7068, Pasadena, CA 91109-7068 (US). (81) Designated States: AU, BR, CA, JP, KR, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i>

(54) Title: PRESSURE-SENSITIVE STRUCTURAL ADHESIVE**(57) Abstract**

A pressure-sensitive structural adhesive is formed by applying a thin layer of pressure-sensitive adhesive (20, 22) to one or both sides of a core layer (18) of partially cured structural adhesive. The pressure-sensitive adhesive provides a tacky surface allowing a temporary bond at room temperature. Upon curing of the construction, the bond properties change to that of a structural adhesive, providing a strong permanent bond. Upon cure, the skin layer or layers of pressure-sensitive adhesive are absorbed into the layer of structural adhesive.

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PRESSURE-SENSITIVE STRUCTURAL ADHESIVE**Field of Invention**

This invention provides a novel pressure-sensitive structural adhesive construction which is normally tacky and forms a pressure-sensitive adhesive bond at room temperature, which bond on heat activation is supplanted by a structural adhesive bond.

Background of the Invention

Structural adhesives such as cyanoacrylate epoxy resins and the like are well-known for providing strong and permanent bonds. However, before curing, such adhesives do not normally provide aggressive adhesive qualities and, therefore, require external aids such as clamping devices to hold the substrates to be bonded together until cure has been completed and a structural bond formed.

Tacky, pressure-sensitive adhesives have been known for many years and have been used in various bonding and fastening applications. They provide a flexible bond and are used in a wide range of applications including adhering labels, decals, and bonding automotive trim parts and name plates to various substrates. However, they show substantially lower strength characteristics compared to a structural adhesive like cured epoxy or cyanoacrylates.

There is a need in the industry to find an adhesive composition which exhibits pressure-sensitive adhesive characteristics useful during assembly and

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which may then be converted by subsequent treatment to a structural adhesive. It is preferred that the subsequent treatment be by thermal means and could even be cured by electromagnetic induction if desired.

1 In use, such adhesive could be provided between
two release layers or as a single tape with
differential release, preferably using a release layer
which is differentially releasable from an adhesive
5 layer bonded to a backing which may also be a release
surface. The adhesive could then be conveniently
applied between the surfaces to be bonded.
Sufficiently firm contact or light pressure established
between the surfaces will cause sufficient adhesion to
10 temporarily hold the assembly. Thereafter, heat is
applied to the composite structure to convert the
intermediate adhesive layer into a structural adhesive
bond.

Applications for such tapes include "hem-flange"
bonding and weld reinforcement in automotive,
15 reinforcing materials for strengthening thin sheet
metal and plastics by forming integral structures with
the original substrate, and bonding of plastics,
precoated metals, SMC, and other advanced materials.

Several attempts were made in the past to come up
20 with a pressure-sensitive adhesive which changes into
a structural adhesive after heat activation.

U.S. Patent 3,326,741 (1967) to Olson discloses a
tacky pressure-sensitive adhesive which on heat
activation achieves a strong permanent bond. The
25 patent discloses a nitrile rubber/epoxy resin blend
with curing agent such as dicyandiamide. However, the
films did not show good cohesive strength at room
temperature which is typically required for a pressure-
sensitive adhesive.

30 U.S. Patent 3,639,500 (1972) to Muny and assigned
to Avery Dennison Corporation discloses a curable
pressure-sensitive adhesive composition containing a

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polyepoxide, a carboxylated diene polymer, and an acrylic ester tackifier which on heat activation provides a structural bond.

U.S. Patent 4,404,246 (1983) to Charbonneau et. al discloses an alkoxyated amino formaldehyde condensate in an acrylic pressure-sensitive adhesive composition as a latent crosslinking agent to improve the cohesive strength after heat activation. However, the material behaves as a highly crosslinked pressure-sensitive adhesive after heat activation and cannot be used for structural bonding.

U.S. Patent 4,452,955 to Boeder discloses an adhesive composition consisting of a polymer dissolved in polymerizable monomer, an accelerator such as organic sulfimides and perfluoroalkylsulfonanilides, and an inhibitor. The adhesive shows pressure-sensitive adhesive properties, and after heat activation, the adhesive demonstrates properties similar to a structural adhesive. However, the properties as detailed in the examples do not show properties of true structural type adhesives, especially in lap shears.

U.S. Patent 4,404,345 (1983) to Janssen also discloses a similar adhesive composition consisting of an adhesive base as the first part and an initiator portion as the second part. Bonding methods are also described. None of the prior art described above teaches or suggests a pressure-sensitive adhesive that can be heat cured to form the strong and permanent bond of a structural adhesive.

Summary of the Invention

This invention provides an adhesive that has the ease of application of a pressure-sensitive adhesive, yet upon cure by application of heat, forms the strong and permanent bond of a structural adhesive. This adhesive is made up of a core layer of a curable permanent structural adhesive having opposed surfaces

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1 with a thin skin of a pressure-sensitive adhesive on
one or both surfaces. The core layer is preferably
made up of either a partially cured B stage structural
adhesive or a blend of curable polymeric material such
as an epoxy with an acrylate resin and hardener which
upon cure will form a structural adhesive. It is this
core layer that forms the strong and permanent bond
after final cure. The preferred embodiment also
5 includes an impact improving elastomer to make the bond
more impact resistant. In the most preferred
embodiment, an impact absorbing agent is included in
the core layer.

10 The skin of pressure-sensitive adhesive that is
applied to one or both opposing surfaces of the core
layer is inherently tacky and provides a temporary bond
between the adhesive and a substrate at room
temperature. This temporary bond created by the
pressure-sensitive adhesive provides a means for
holding two substrates together during the cure of the
15 core layer structural adhesive. The core layer
structural adhesive is cured by heating the entire
assembly made up of the two substrates and the
adhesive. Once the heat-cure process has been
completed, a strong and permanent bond results. During
20 cure, the skin or skins of pressure-sensitive adhesive
are absorbed, presumably blending into the core layer
structural adhesive.

25 Such an adhesive is useful in that it eliminates
the need to clamp or otherwise hold substrates to be
bonded together during heat cure. The skin or skins of
pressure-sensitive adhesive act in place of clamps by
providing a temporary bond that lasts long enough to
enable the structural adhesive to be cured.

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Brief Description of the Drawings

FIG. 1 is a cross section of one embodiment of the invention.

FIG. 2 is a cross section of a second embodiment of the invention.

FIG. 3 is a cross section of a third embodiment of the invention.

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Detailed Description

1 This invention provides a novel pressure-sensitive
adhesive construction which is normally tacky and forms
a pressure-sensitive adhesive bond at room temperature
and which on heat activation is supplanted by a
structural adhesive bond. This is achieved by
distributing on one or both sides of a core layer of
structural adhesive a thin skin of a pressure-sensitive
adhesive. If the pressure-sensitive adhesive is
5 applied to only one side of the core layer, the other
side of the core layer can be prebonded to a backing or
face stock. The skin or skins of pressure-sensitive
adhesive can be either continuous or discontinuous
layers. Various different pressure-sensitive adhesives
10 can be used for the skin layer or layers. For example,
either acrylic- or rubber-based pressure-sensitive
adhesive may be used. Additionally, the
pressure-sensitive adhesive can be either one that is
inherently tacky or one that requires addition of a
15 tackifier prior to bonding. The core layer is made up
of various different structural adhesives such as a
partially cured B stage structural adhesive or a blend
of epoxy with an acrylate ester resin and hardener.
While the skin or skins of pressure-sensitive adhesive
20 provide the initial tack, these layers are absorbed
into the core layer by heat activation. This unique
feature provides a pressure-sensitive adhesive with
structural adhesive properties after heat activation.

Referring to the figures, three different
25 embodiments of the invention are illustrated in FIGS.
1-3. In FIG. 1, a cross section of the adhesive is
shown in which a core layer of structural adhesive is
sandwiched between two continuous skin layers of
pressure-sensitive adhesive 12 and 14. The
30 illustration also shows a suitable release liner 16
which protects the pressure-sensitive adhesive and

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prevents the adhesive from inadvertent bonding prior to use.

FIG. 2 is a cross section of a second embodiment of the invention in which the core layer of structural adhesive 18 is sandwiched between two discontinuous skin layers of pressure-sensitive adhesive 20 and 22. These discontinuous skin layers can take the form of stripes, dots or various other patterns of pressure-sensitive adhesive. The release liner is illustrated as 24.

FIG. 3 is a cross section of a third embodiment of the invention. In this embodiment, the core layer of structural adhesive 26 is prebonded to substrate 28. A skin layer of pressure-sensitive adhesive 30 is found on the opposite side of the core layer. Release liner 32 protects the adhesive and prevents inadvertent bonding prior to use.

This invention is useful for various applications where a strong bond between two substrates or between a backing and a substrate is desirable but difficult to achieve because the substrates are difficult to hold together while the adhesive cures. Applications for such adhesives are numerous. In the automobile manufacturing industry, such adhesives would be useful for "hem-flange" bonding and weld reinforcement by providing a temporary bond which would become permanent during the paint-bake step in manufacturing. The adhesive would also be useful in the aerospace and other industries by providing a means of easily forming integral bonds between plastics, metals and various advanced materials.

The core layer can be comprised of an epoxy resin, a polymer resin and a hardener. Typical examples of epoxy resins include Epon 828, Epon 826, Epon 836 and the like available from Shell Chemical Co., and are characterized by the presence of epoxide functionality. The resins at room temperature could be a liquid of low

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molecular weight or solid resins which are higher in molecular weight. Blends of several epoxy resins of different structures, molecular weight and epoxy functionality could be used to achieve the desired balance of properties for the core layer.

1 The polymer resin is preferably an acrylic ester resin like polymethyl methacrylate (PMMA) and PMMA copolymers and the like. For example Acryloid Resin A 21, B 667 etc. manufactured by Rohm and Haas can be
5 used for this application. The acrylic ester resins could be based on methyl methacrylate, butyl acrylate, and isobutyl methacrylate and the like. Other polymers or polymer blends can also be used.

10 Typical examples of hardeners include boron trifluoride or trichloride amine complexes like BF_3 : monoethyl amine, blocked amines like HT 9506 (produced by Ciba Geigy), or Dicyandiamide (Dicy) and the like. Mixtures of a blend of hardeners could also be used.
15 Other types of epoxy hardeners could also be used if they provide the desired stability and efficient cure during the heat activation. The hardeners would preferably be incompatible with the resin or inactive at room temperature but if compatible, the hardeners could also be encapsulated in a heat or pressure-
20 sensitive polymer shell.

 The core layer can also be comprised of a partially cured B stage epoxy resin. A typical example of such a resin is the FM-73 type manufactured by American Cyanamide. Additional fillers, modifying
25 additives, fibers, and the like could be added to the core layer to improve the strength or modify the properties of the core layer. Due to brittleness of resins without fillers, the preferred embodiment includes fillers. Low density additives like
30 microballoons could be incorporated if desired. Electromagnetic materials, particulate magnetizable iron, cobalt, nickel, alloys of nickel and iron, alloys

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of nickel and chromium, inorganic oxides of iron, inorganic oxides of nickel and the like could be included to make the adhesive induction curable. The material could also be formulated for UHF, radio frequency, or microwave curing if desired.

1 The skin layer or layers of pressure-sensitive adhesive can be comprised of an acrylic pressure-sensitive adhesive such as Polytex 7000 which is
5 produced by Avery Chemical, Division of Avery Dennison Corporation. This is a high performance pressure-sensitive adhesive within the scope of the claims of U.S. Patent 4,812,541 to Mallia et al., incorporated
10 herein by reference. This high performance pressure-sensitive adhesive provides unusually high adhesion to high energy surfaces such as aluminum and stainless steel due to synergistic combination of a glycidyl monomer and an N-vinyl lactam monomer.

15 The skin layer or layers of pressure-sensitive adhesive can also be comprised of an elastomeric pressure-sensitive adhesive. Curable elastomeric pressure-sensitive adhesives are disclosed in U.S. Patent 4,948,825 to Sasaki, incorporated herein by
20 reference. These curable elastomeric pressure-sensitive adhesives incorporate organic additives to reduce the energy requirements of a pressure-sensitive adhesive that is cured by actinic radiation.

25 Various other pressure-sensitive adhesives with different properties can be used for the skin layer as well. A permanent pressure-sensitive adhesive can be used, or if repositioning of the substrates to be bonded is desired prior to final cure, a removable pressure-sensitive adhesive can be used. A repositionable pressure-sensitive adhesive is disclosed
30 in U.S. Patent Application Serial No. 741,556 to Mallia et al., incorporated herein by reference. This application discloses a repositionable pressure-

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sensitive adhesive that has adhesive characteristics that vary depending on the application pressure. Furthermore, an inherently tacky pressure-sensitive adhesive can be used, or one that requires the addition of a tackifier prior to bonding.

1 The impact resistance of the pressure-sensitive structural adhesives disclosed in this specification tends to be low. Impact resistance can be improved by the addition of fillers, modifying additives, fibers or
5 the like. These additives are included in the core layer at levels which provide satisfactory impact resistance and reduced brittleness. Since the skin layer of pressure-sensitive adhesive is absorbed into the core layer during cure, the impact modifier can
10 also be added to the pressure-sensitive adhesive as well as the core layer with equivalent results.

 While numerous options are available for the skin layers of pressure-sensitive adhesive that make up this pressure-sensitive structural adhesive, the preferred
15 embodiment uses an acrylic pressure-sensitive adhesive that includes a compatible impact-improving elastomer.

 Various release layers are available which may be applied to the adhesive and are useful in protecting the skin layer or layers of pressure-sensitive adhesive from inadvertently bonding prior to use. Suitable
20 release layers are described in some detail in Chapter 23 of the Handbook of Pressure Sensitive Adhesive Technology, 2d Ed., edited by Donatas Satus, and incorporated herein by reference. If skin layers of
25 pressure-sensitive adhesive are used on both sides of the core layer of structural adhesive, then release layers can be applied to both sides of the adhesive. These two release layers would preferably be differentially releasable from the adhesive layer to
30 provide additional convenience in application.

 Following are examples which more specifically illustrate the invention. In these examples, the 180°

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1 peel adhesion is measured as described in PSTC-1, using
2 mil Mylar as face material and stainless steel
substrate. The shear adhesion failure temperature
(SAFT) is measured by applying the adhesive to a 2 mil
aluminum strip and bonding it to stainless steel with
a 2.54 cm² overlap. A 1.0 kg load is attached on one
end and the temperature is raised at 0.5°C per minute
until the adhesive fails in shear. The temperature at
which the adhesive fails is regarded as the SAFT
5 temperature.

Lap shear is determined by applying the adhesive
film to a 0.64 to 1.01 mm thick by 2.54 cm strip of
steel that is subsequently bonded to a second strip of
steel with the same dimensions. A 1.27 cm overlap is
10 maintained between the steel strips for a total bond
area of 1.27 cm by 2.54 cm. A 0.5 mm bond thickness
for the structural adhesive is maintained by using 0.5
mm spacer bars placed between the steel strips. An
external force is applied to pull the strips apart, and
15 the lap shear is measured as the force at which the
bond breaks divided by the bond area. Lap shear is
reported in pressure units as psi or MPa. Since the
bonded steel strips are offset in nature, the measured
bond failure is not purely due to shear stress, but
20 includes cleavage and peel stresses as well. This
determination of lap shear follows general engineering
standards of the automobile industry.

Example 1:

25 A core layer was prepared by mixing 48 parts of
Acryloid B66T (51% in Toluene), 16 part of Epon 828 and
8 parts of HT9506. The mixture was coated onto a
Teflon FEP film to give a coat weight of 75 g/m². This
30 was first dried for 15-20 minutes at room temperature
and then at 70°C for 15 minutes.

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1 A skin layer of an acrylic pressure-sensitive adhesive was prepared by solvent coating 15 g/m² of Polytex 7000 onto a Teflon FEP film and drying at 70°C for 15 minutes. The skin layer was then laminated on both sides of the core layer to prepare a sandwich construction.

5 This film gave a 180° peel adhesion of 526 N/m, shear adhesion of 441 minutes, shear adhesion failure temperature of 140°F and lap shear of 189 psi (1.3 MPa). After baking this film at 200°C for 30 minutes, it gave a lap shear of 1000 psi+ (6.89 MPa).

10 Example 2:

A "B" stage epoxy resin (FM-73) from American Cyanamide was used as the middle core layer.

15 A skin layer of pressure-sensitive adhesive (Polytex 7000) was laminated to each side of the core layer at 15 g/m² coat weight.

20 This film gave a lap shear of 37 psi. After baking at 200°C, this film gave a lap shear of 1000 + psi.

Example 3:

25 A core layer was prepared by mixing 66 parts of Acryloid B21 (30% in Toluene), 39 parts of Epon 828 and 12 parts of HT9506. The mixture was coated onto a Teflon FEP film to give a coat weight of 75 g/m². This was first dried for 15-20 minutes at room temperature and then at 70°C for 15 minutes.

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1 A skin layer of an acrylic pressure-sensitive adhesive was prepared by coating 15 g/m² of Polytex 7000 onto a Teflon FEP film and drying at 70°C for 15 minutes. The skin layer was then laminated on both sides of the core layer to prepare a sandwich construction.

5 This film gave a 180° peel adhesion of 526 N/m, shear adhesion of 2000 minutes+, shear adhesion failure temperature of 183°F and lap shear of 174 psi (1.23 MPa). After baking at 200°C for 30 minutes, this film gave a lap shear of 1000 psi+ (6.89 MPa) typical of a structural adhesive.

10 Example 4:

15 A middle core layer was prepared by blending 227 parts of Araldite GY 6010, an unmodified liquid epoxy resin (manufactured by CIBA GEIGY), and 95 parts of Acryloid B66, in a Brabender Plasticorder using the roller mixer at 140° C until the mixture was homogenous. The temperature of the mix was reduced to 20 45-50°C by cooling and 32 parts of Dicyanex 200-X, a solid curing agent, (manufactured by American Cyanamide) was mixed into this mixture. This blend was then extruded as a sheet, 3 to 6 mil in thickness using 25 the Brabender extruder. The die temperature was maintained at about 50 to 60°C. Two thin (15 g/m²) acrylic pressure-sensitive adhesive layers were 30 laminated on either side of this core.

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This film laminated to 2 mil Mylar showed a peel adhesion of 1400 N/m on stainless steel substrate, room temperature shear of 255 minutes and SAFT of 45°C. The sample showed a lap shear of 105 psi (0.72 MPa). The lap shear of the sample after baking at 200°C for 30 minutes increased to 2800 psi (19.29 MPa).

Example 5:

A core layer was prepared by blending 300 parts of Acryloid A-21, a solid acrylic copolymer resin, heated to 175°C with 400 parts of Araldite GY-6010, an unmodified liquid epoxy resin, until homogenous, raising the temperature to 215°C and adding 44 parts of glass fiber (965-57) and mixing for about 30 minutes. The mixture was cooled to 90°C and 50 parts Hycar 1300 x 13, an acrylic copolymer, were added then the blend was mixed for an additional 30 minutes. Finally 70 parts of Dicyanex 200-X, a solid curing agent, were added and mixed for another 30 minutes. The mixture was coated onto a Teflon FEP film to give a coat weight of 550 g/m².

Two skin layers of Polytex 7000 adhesive film were laminated to either side of the core layer to give a coat weight of 17 g/m². This sample gave a 180° peel adhesion of 1020 N/m. The lap shear after baking at 196°C for 30 minutes was 1970 psi.

Example 6:

A core layer was prepared as in Example 5 to a coat weight of 625 g/m². Skin layers of I-406 were

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laminated to either side of the core layer to give a coat weight of 10.6 g/m².

The film gave a 180° peel adhesion of 875 N/m. After baking at 196°C for 30 minutes, the sample gave a lap shear of 1400 psi.

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WHAT IS CLAIMED IS:

1. A pressure-sensitive structural adhesive comprising a core layer of a curable structural adhesive providing opposed surfaces and having on at least one surface thereof a separate continuous or discontinuous layer of a tacky pressure-sensitive adhesive, the pressure-sensitive adhesive providing an initial adhesion between a substrate to be bonded by the cured structural adhesive, said pressure-sensitive adhesive layer being absorbed by the core layer of curable structural adhesive when a permanent thermoset bond to such substrate is formed by thermal cure of the curable structural adhesive.

2. The pressure-sensitive structural adhesive as claimed in claim 1 in which there is present an impact improving resin which is compatible with the structural adhesive and included in the core layer, the pressure-sensitive adhesive layer or the core layer and pressure-sensitive adhesive layer.

3. The pressure-sensitive structural adhesive as claimed in claim 2 in which the compatible impact improving resin is an elastomer.

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4. The pressure-sensitive structural adhesive as claimed in any of claims 1 to 3 in which the core layer is comprised of partially cured B-stage epoxy resin.

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5. The pressure-sensitive structural adhesive as claimed in any of claims 1 to 4 in which the core layer of curable structural adhesive is comprised of a blend
5 of an epoxy resin with acrylate resin and hardener.

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6. The pressure-sensitive structural adhesive as claimed in any of claims 1 to 5 in which the pressure-sensitive structural adhesive is on both surfaces of
10 the core layer and the pressure-sensitive adhesive layers bond two substrates together until the core layer of curable structural adhesive is cured by
15 application of heat.

15

7. The pressure-sensitive structural adhesive as
20 claimed in any one of claims 1 to 6 in which electromagnetic materials are blended into the core layer of curable structural adhesive and the structural adhesive is themally cured by electromagnetic induction
25 heating.

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Fig. 1

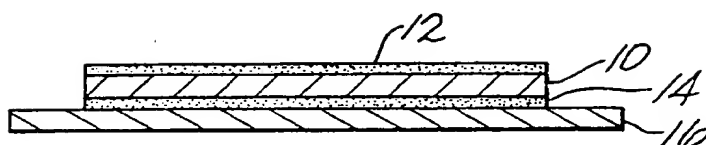


Fig. 2

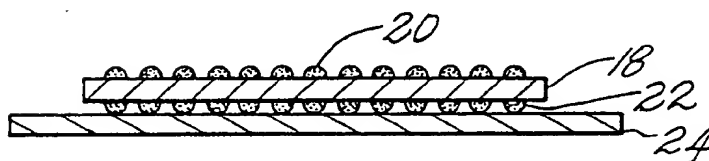
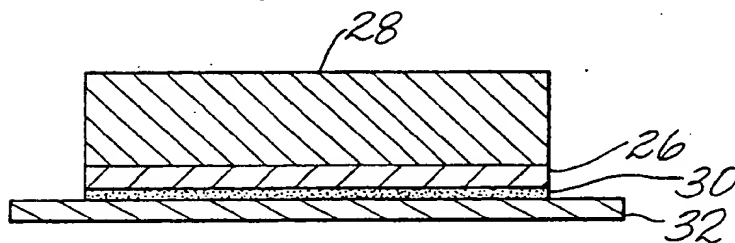


Fig. 3



INTERNATIONAL SEARCH REPORT

International application No.
PCT/US93/09996

A. CLASSIFICATION OF SUBJECT MATTER IPC(5) :B32B 7/10 US CL :428/200, 343, 344, 346, 347, 354, 355 According to International Patent Classification (IPC) or to both national classification and IPC				
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) U.S. : 428/200, 343, 344, 346, 347, 354, 355 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)				
C. DOCUMENTS CONSIDERED TO BE RELEVANT				
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.		
Y	US, A, 3,639,500 (MUNY) 01 FEBRUARY 1972. See entire document.	1-4		
Y	US, A, 3,326,741 (OLSON) 20 JUNE 1967. See entire document.	1-4		
Y	US, A, 4,948,825 (SASAKI) 14 AUGUST 1990. See column 2, lines 25-36.	2-4		
<input type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family annex.				
<table border="0"> <tr> <td> * Special categories of cited documents: "A" document defining the general state of the art which is not considered to be part of particular relevance "E" earlier document published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed </td> <td> "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "Z" document member of the same patent family </td> </tr> </table>			* Special categories of cited documents: "A" document defining the general state of the art which is not considered to be part of particular relevance "E" earlier document published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "Z" document member of the same patent family
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Date of the actual completion of the international search 01 December 1993		Date of mailing of the international search report JAN 03 1994		
Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. NOT APPLICABLE		Authorized officer CHRISTOPHER BROWN Telephone No. (703) 308-2351		

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US93/09996

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☒ Claims Nos.: 5-7
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐

The additional search fees were accompanied by the applicant's protest.

☐

No protest accompanied the payment of additional search fees.

Form PCT/ISA/210 (continuation of first sheet(1))(July 1992)*